Nuclear Magnetic Relaxation in the Haldane-Gap Antiferromagnet $Ni(C_2H_8N_2)_2NO_2(ClO_4)$

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A new theory is proposed to interpret nuclear spin-lattice relaxation-time (T_1) measurements on the spin-1 quasi-one-dimensional Heisenberg antiferromagnet $Ni(C_2H_8N_2)_2NO_2(ClO_4)$ (NENP). While Sagi and Affleck pioneeringly discussed this subject in terms of field-theoretical languages, there is no theoretical attempt yet to explicitly simulate the novel observations of T_1^{-1} reported by Fujiwara *et al.*. By means of modified spin waves, we solve *the minimum of T_1^{-1} as a function of an applied field*, pending for the past decade.

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Predicting a striking contrast between integer- and half-odd-integer-spin one-dimensional Heisenberg antiferromagnets, Haldane [1,2] sparked renewed interest in low-dimensional quantum magnetism. The Haldane gap, that is, a magnetic excitation gap immediately above the ground state, was not only calculated by various numerical tools [3–8] but indeed observed in spin-1 quasi-one-dimensional Heisenberg antiferromagnets such as $CsNiCl_3$ [9] and $Ni(C_2H_8N_2)_2NO_2(ClO_4)$ (NENP) [10]. The valence-bond-solid model [11,12] significantly contributed toward understanding novel features of the Haldane massive phase such as fractional spins induced on the boundaries [13,14], a string order hidden in the ground state [15,16] and magnon excitations against the hidden order [17–20]. The nonlinear-σ-model quantum field theory [21,22] skillfully visualized the competition between massive and massless phases, while a generalized Lieb-Schultz-Mattis theorem [23,24] gave a criterion for the gap formation in a magnetic field.

Recent progress in the experimental studies also deserves special mention. Not only the single-magnon dispersion relation [25] but also the two-magnon continuum [3,26] was directly observed by inelastic-neutron-scattering measurements on NENP [27] and $CsNiCl_3$ [28]. An applied magnetic field may destroy the Haldane gap and bring back magnetism to the system. Such a field-induced long-range order was indeed realized in a nickel compound $Ni(C_5H_{14}N_2)_2N_3(PF_6)$ [29]. Another family of linear-chain nickelates of general formula R_2BaNiO_5 (R = rare earth or Y) [30] exhibited a novel scenario of one- to three-dimensional crossover [31,32]. When the nonmagnetic Y^{3+} ions are substituted by other magnetic rare-earth ions in Y_2BaNiO_5 with a disordered ground state, there appears a three-dimensional long-range order, while the one-dimensional gapped excitations persist both above and below the Néel temperature [33–35].

Nuclear spin-lattice relaxation time (T_1) has also been measured on Haldane-gap antiferromagnets. The field (H) dependence of T_1^{-1} is of particular interest at both high and low temperatures. The high-temperature relaxation rate has been discussed in the context of transport properties. Takigawa *et al.* [36] measured the high-temperature relaxation rate of ^{31}P and ^{51}V nuclei of $AgVP_2S_6$, which is also an ideal spin-1 Haldane-gap antiferromagnet, and observed the diffusive dynamics $T_1^{-1} \propto H^{-1/2}$ [37]. There is a hot argument [38–41] whether the spin transport in quantum spin-gapped antiferromagnets should be diffusive or ballistic at finite temperatures. On the other hand, Gaveau *et al.* [42] and Fujiwara *et al.* [43,44] measured the low-temperature relaxation rate of ^1H nuclei of NENP. As an applied field increases, the first excited state moves down and then crosses the ground-state energy level. Indeed T_1^{-1} reaches a peak near the critical field $H_c \equiv \Delta_0/g\mu_B$ at every temperature, but it is not a monotonically increasing function of H, at low temperatures $T \lesssim \Delta_0/k_B$ in particular. Sagi and Affleck [45] formulated the nuclear magnetic resonance in Haldane-gap antiferromagnets in terms of field-theoretical languages. However, few investigations have followed their pioneering argument and there is no attempt yet to explicitly fit a theory for the above observations.

In such circumstances, we revisit the low-temperature nuclear magnetic relaxation in Haldane-gap antiferromagnets with particular emphasis on its field dependence. Excluding any phenomenological assumption from our argument, we calculate the nuclear spin-lattice relaxation rate by means of modified spin waves. A new theory claims that *as an applied field increases, T_1^{-1} should initially decrease logarithmically and then increase exponentially*, well explaining experimental observations.

We employ the spin-1 one-dimensional Heisenberg Hamiltonian

$$\mathcal{H} = J \sum_{l=1}^{L} \boldsymbol{S}_l \cdot \boldsymbol{S}_{l+1} - g\mu_B H \sum_{l=1}^{L} S_l^z \,. \tag{1}$$

In order to illuminate the essential relaxation mechanism in spin-gapped antiferromagnets as analytically as possible, we do not take any anisotropy into consideration in this letter. Magnetic anisotropy quantitatively affects the gap amplitude but has no qualitative effect on the whole scenario. Scaling temperature and an applied field by the Haldane gap, we present a universal theory. Quantitative

refinement of the final product in the presence of single-ion and orthorhombic anisotropy terms will be considered elsewhere. Introducing the Holstein-Primakoff bosonic operators

$$\begin{aligned} S^+_{2n-1} &= \sqrt{2S - a^\dagger_n a_n}\; a_n\,, \;\; S^z_{2n-1} = S - a^\dagger_n a_n\,, \\ S^+_{2n} &= b^\dagger_n \sqrt{2S - b^\dagger_n b_n}\;, \qquad S^z_{2n} = -S + b^\dagger_n b_n\,, \end{aligned} \tag{2}$$

and retaining only bilinear terms of them, we rewrite the Hamiltonian as

$$\begin{aligned} \mathcal{H} &= -2JS^2N \\ &+(2JS + g\mu_{\rm B}H)\sum_{n=1}^{N} a^\dagger_n a_n + (2JS - g\mu_{\rm B}H)\sum_{n=1}^{N} b^\dagger_n b_n \\ &+JS\sum_{n=1}^{N}\left(a^\dagger_n b^\dagger_n + a_n b_n + b^\dagger_n a^\dagger_{n+1} + b_n a_{n+1}\right)\,, \end{aligned} \tag{3}$$

where $N \equiv L/2$. In order to preserve the up-down symmetry, or the sublattice symmetry, we optimize the spin-wave distribution functions constraining the total staggered magnetization to be zero [46–49]:

$$\sum_n \left(a^\dagger_n a_n + b^\dagger_n b_n\right) = 2NS\,. \tag{4}$$

The constraint is enforced by introducing a Lagrange multiplier and diagonalizing an effective Hamiltonian

$$\widetilde{\mathcal{H}} = \mathcal{H} + 2J\lambda\sum_n \left(a^\dagger_n a_n + b^\dagger_n b_n\right)\,. \tag{5}$$

Via the Bogoliubov transformation

$$\begin{aligned} \frac{1}{\sqrt{N}}\sum_n \mathrm{e}^{\mathrm{i}k(2n-1/2)} a_n &= \alpha_k \cosh\theta_k - \beta^\dagger_k \sinh\theta_k\,, \\ \frac{1}{\sqrt{N}}\sum_n \mathrm{e}^{-\mathrm{i}k(2n+1/2)} b_n &= \beta_k \cosh\theta_k - \alpha^\dagger_k \sinh\theta_k\,, \end{aligned} \tag{6}$$

with $\tanh 2\theta_k = S\cos k/(S+\lambda)$, we reach the spin-wave Hamiltonian

$$\begin{aligned} \mathcal{H} &= -2JS^2N - 2J(S+\lambda)N + J\sum_k \omega_k \\ &+J\sum_k\left(\omega^-_k \alpha^\dagger_k\alpha_k + \omega^+_k\beta^\dagger_k\beta_k\right)\,, \end{aligned} \tag{7}$$

where

$$\omega^\pm_k \mp g\mu_{\rm B}H/J = 2\sqrt{(S+\lambda)^2 - S^2\cos^2 k} \equiv \omega_k\,. \tag{8}$$

Minimization of the free energy gives the optimum distribution functions as $\bar{n}^\pm_k = (\mathrm{e}^{J\omega^\pm_k/k_{\rm B}T} - 1)^{-1}$ and λ is then determined through

$$\sum_k \left(\bar{n}^-_k + \bar{n}^+_k + 1\right)\cosh 2\theta_k = (2S+1)N\,. \tag{9}$$

Now we calculate the relaxation rate in terms of the modified spin waves. Considering the electronic-nuclear energy-conservation requirement, the Raman scattering predominates in spin-gapped antiferromagnets. The Raman relaxation rate is generally given by

$$\begin{aligned} \frac{1}{T_1} &= \frac{4\pi(g\mu_{\rm B}\hbar\gamma_{\rm N})^2}{\hbar\sum_i \mathrm{e}^{-E_i/k_{\rm B}T}}\sum_{i,j}\mathrm{e}^{-E_i/k_{\rm B}T} \\ &\times\left|\langle j|\textstyle\sum_l A_l S^z_l|i\rangle\right|^2\delta(E_j - E_i - \hbar\omega_{\rm N})\,, \end{aligned} \tag{10}$$

where A_l is the dipolar coupling constants between the nuclear and electronic spins in the lth site, $\omega_{\rm N} \equiv \gamma_{\rm N}H$ is the Larmor frequency of the nuclei with $\gamma_{\rm N}$ being the gyromagnetic ratio, and the summation $\sum_i$ is taken over all the electronic eigenstates $|i\rangle$ with energy E_i. By means of the modified spin waves, eq. (10) is rewritten as

$$\begin{aligned} \frac{1}{T_1} &= \frac{4\pi(g\mu_{\rm B}\hbar\gamma_{\rm N})^2}{\hbar N^2\sum_i \mathrm{e}^{-E_i/k_{\rm B}T}}\sum_{i,j}\mathrm{e}^{-E_i/k_{\rm B}T}\delta(E_j - E_i - \hbar\omega_{\rm N}) \\ &\times\Big|\langle j|\textstyle\sum_{k,k'} A_{k'-k}\big[\big(\alpha^\dagger_{k'}\alpha_k - \beta^\dagger_{k'}\beta_k\big)\cosh\theta_{k'}\cosh\theta_k \\ &-\big(\alpha_{k'}\alpha^\dagger_k - \beta_{k'}\beta^\dagger_k\big)\sinh\theta_{k'}\sinh\theta_k\big]|i\rangle\Big|^2\,, \end{aligned} \tag{11}$$

where $A_q = \sum_l \mathrm{e}^{\mathrm{i}ql}A_l$. The Fourier components of the hyperfine coupling constant exhibit little momentum dependence when the nuclei take unsymmetrical positions in the crystal, which is the case with the protons in NENP [44]. Hence we assume in the following that $A_q \simeq A_{q=0} \equiv A$. Due to the significant difference between the electronic and nuclear energy scales ($\hbar\omega_{\rm N} \lesssim 10^{-5}J$), eq. (11) ends in

$$\frac{1}{T_1} = \frac{4(g\mu_{\rm B}\hbar\gamma_{\rm N}A)^2}{\pi\hbar}\int_{-\pi/2}^{\pi/2}\frac{\sum_{\sigma=\pm}\bar{n}^\sigma_k(\bar{n}^\sigma_k+1)}{\sqrt{v^2k^2 + v\hbar\omega_{\rm N}}}\mathrm{d}k\,, \tag{12}$$

where assuming moderate temperatures $k_{\rm B}T \ll \omega^-_{k=\pi/2} = 2J(S+\lambda) - g\mu_{\rm B}H$, we have approximated the dispersion relations as

$$J\omega^\pm_k \simeq \Delta + vk^2 \pm g\mu_{\rm B}H\,, \tag{13}$$

with

$$\Delta = 2J\sqrt{\lambda(2S+\lambda)}\,, \;\; v = \frac{JS^2}{\sqrt{\lambda(2S+\lambda)}}\,. \tag{14}$$

Equation (12) claims that an applied field produces two distinct effects on T_1^{-1}, one of which originates from the Zeeman energy and appears in $\bar{n}^\sigma_k$, while the other of which appears via the nuclear spins giving the characteristic weight $(v^2k^2 + v\hbar\omega_{\rm N})^{-1/2}$ to the electronic transition rate $\bar{n}^\sigma_k(\bar{n}^\sigma_k+1)$. The field effect on the nuclear spins escapes observation in critical spin chains with a linear dispersion at small momenta. The prefactor $(v^2k^2 + v\hbar\omega_{\rm N})^{-1/2}$ is the consequence of the nature of the delta function,

$$\delta[f(x)] = \sum_i \frac{\delta(x - x_i)}{|f'(x_i)|}\,, \tag{15}$$

where x_i is a zero point of an arbitrary regular function $f(x)$, and therefore generally arises from quadratic dispersion relations of the relevant electronic excitations, which are the case with ferromagnets [50–52] as well as spin-gapped antiferromagnets.

Let us fit eq. (12) for the proton spin-lattice relaxation-time measurements on NENP [44]. Although Δ and v, given in eq. (14), depend on temperature in principle, here we fix $(v^2k^2 + v\hbar\omega_{\rm N})^{-1/2}$ to its zero-temperature value in the integration (12), which is well justified for $k_{\rm B}T \lesssim \Delta - g\mu_{\rm B}H$ and allows us to inquire further into eq. (12) analytically. We compare the calculations with the observations in Fig. 1. Assuming that $g = 2$ and $J/k_{\rm B} = 55\,{\rm K}$ [44], we have set A equal to $0.024\,{\rm \AA}^{-3}$, which suggests the distance between the interacting proton and electron spins being about $3.5\,{\rm \AA}$ and is consistent very well with the structural analysis [53]. Under the present parametrization, the lowest excitation gap is given by $\Delta(T=0)/k_{\rm B} \equiv \Delta_0/k_{\rm B} \simeq 4.0\,{\rm K}$, which is somewhat smaller than that of NENP, $12.8\,{\rm K}$ [44]. However, the scaled function $\Delta(T)/\Delta_0$ well reproduces the upward behavior of the Haldane-gap mode as a function of temperature [49].

With increasing field, the relaxation rate first decreases moderately and then increases much more rapidly, at low temperatures in particular. Although we cannot calculate beyond the critical field $H_{\rm c} \equiv \Delta_0/g\mu_{\rm B} \simeq 9.5\,{\rm T}$ on the present formulation, our theory well reproduces the observations over a wide field range. For $k_{\rm B}T \lesssim \Delta - g\mu_{\rm B}H$, the distribution functions may be approximated as $\bar{n}_k^{\pm}(\bar{n}_k^{\pm}+1) \simeq {\rm e}^{-\omega_k^{\pm}/k_{\rm B}T}$ and therefore eq. (12) can further be calculated as

$$\frac{1}{T_1} \simeq \frac{8(g\mu_{\rm B}\hbar\gamma_{\rm N}A)^2}{\pi\hbar v}{\rm e}^{-\Delta_0/k_{\rm B}T}\cosh\Bigl(\frac{g\mu_{\rm B}H}{k_{\rm B}T}\Bigr)K_0\Bigl(\frac{\hbar\omega_{\rm N}}{2k_{\rm B}T}\Bigr)\,, \tag{16}$$

where K_0 is the modified Bessel function of the second kind. Provided $\hbar\omega_{\rm N} \ll k_{\rm B}T \lesssim \Delta - g\mu_{\rm B}H$, we further reach

$$\frac{1}{T_1} \simeq \frac{8(g\mu_{\rm B}\hbar\gamma_{\rm N}A)^2}{\pi\hbar v}{\rm e}^{-\Delta_0/k_{\rm B}T}\cosh\Bigl(\frac{g\mu_{\rm B}H}{k_{\rm B}T}\Bigr) \times\left[0.80908 - \ln\Bigl(\frac{\hbar\omega_{\rm N}}{k_{\rm B}T}\Bigr)\right]. \tag{17}$$

Thus, *as H increases, T_1^{-1} should initially decrease logarithmically and then increase exponentially.* Figure 2 suggests that the initial decrease of T_1^{-1} turns from $T_1^{-1} \propto -\ln H$ to $T_1^{-1} \propto H^{-1/2}$ with decreasing temperature. The momentum distribution functions $\bar{n}_k^{\pm}$ are peaked at $k = 0$ and their peaks are sharpened as T decreases. $\bar{n}_k^{\pm}$ behaves as $\delta(k)$ in the low-temperature limit. When we replace $\bar{n}_k^{\pm}$ by $\delta(k)$, eq. (12) gives a $H^{-1/2}$-linear field dependence of T_1^{-1}.

The temperature dependence is mainly described by the term ${\rm e}^{-(\Delta_0 - g\mu_{\rm B}H)/k_{\rm B}T}$ but further decorated due to the temperature-dependent energy spectrum. The inelastic-neutron-scattering peak position of the lowest-energy mode exhibits an upward behavior with increasing temperature, for $k_{\rm B}T \gtrsim \Delta_0/2$ in particular [54–56], where the slope of $\ln T_1^{-1}$ to T^{-1} correspondingly increases with increasing temperature.

The nuclear magnetic relaxation in the Haldane-gap antiferromagnet NENP has been interpreted in terms of a modified spin-wave theory. The field dependence of T_1^{-1} was analyzed in detail and *the minimum of T_1^{-1} as a function of H, pending for the past decade, was solved.* We consider that such a field dependence of T_1^{-1} is qualitatively common to spin-gapped antiferromagnets. We encourage low-temperature T_1 measurements on related materials such as the ferromagnetic-antiferromagnetic bond-alternating compound $(CH_3)_2CHNH_3CuCl_3$ [57] and the two-leg ladder antiferromagnet $SrCu_2O_3$ [58].

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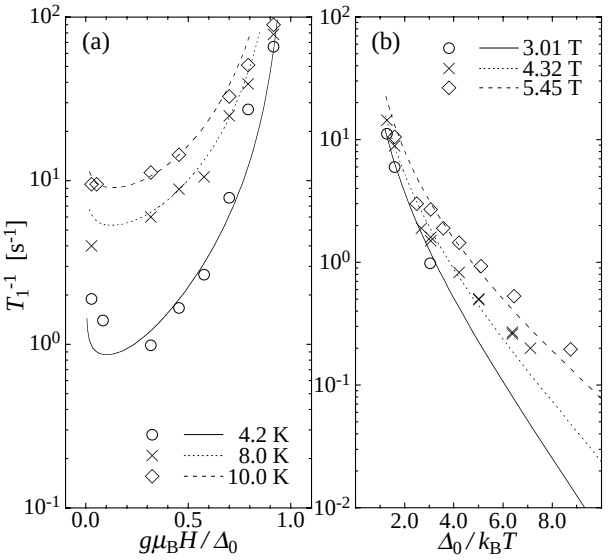

FIG. 1. Dependences of the nuclear spin-lattice relaxation rate on a field parallel to the chain (a) and temperature (b) in NENP (symbols) [44] are compared with the modified spin-wave calculations (lines), where the field and temperature are scaled by the lowest excitation gap Δ_0.

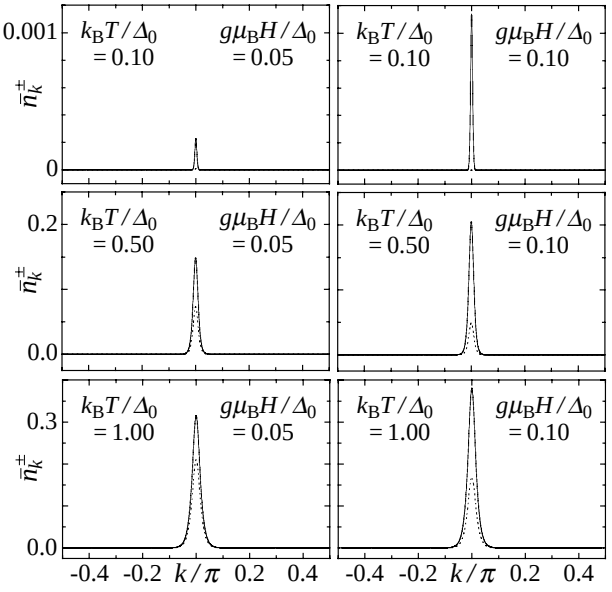

FIG. 2. The momentum distribution functions $\bar{n}_k^+$ (dotted lines) and $\bar{n}_k^-$ (solid lines).